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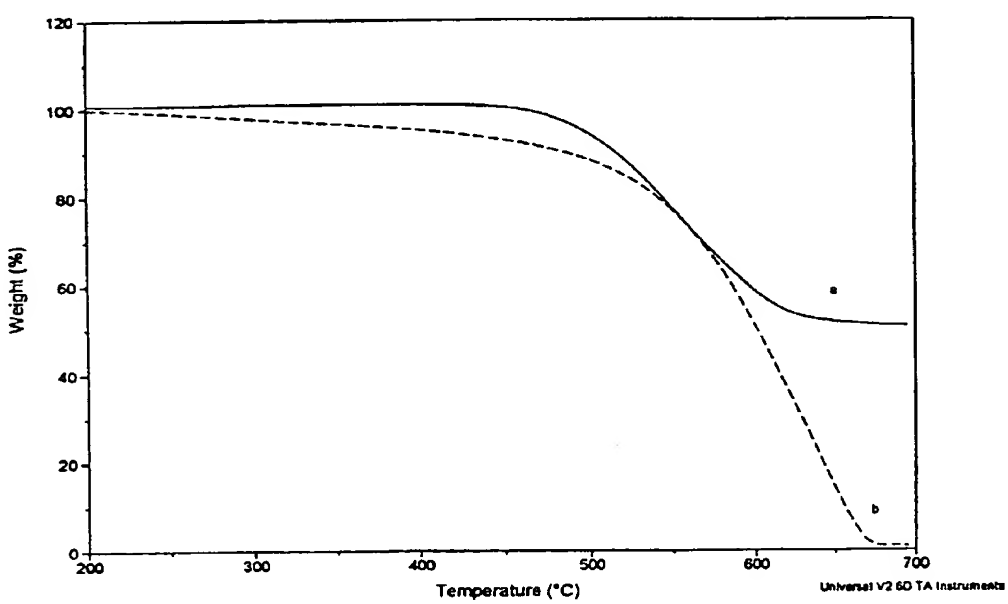
(54) Title: HIGH YIELD VAPOR PHASE DEPOSITION METHOD FOR LARGE SCALE SINGLE WALLED CARBON NANOTUBE PREPARATION



Typical TGA curves for (a) as-prepared and (b) purified SWNT materials in air.

(57) Abstract: An improved vapor phase deposition method for preparation of single walled carbon nanotubes on an aerogel supported metal catalyst. The total yield of SWCNTs often is at least about 100 %, based the weight of the catalyst, for a reaction time of at least about 30 minutes.

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Description

HIGH YIELD VAPOR PHASE DEPOSITION METHOD FOR LARGE SCALE SINGLE WALLED CARBON NANOTUBE PREPARATION

Government Interest

This work is in part supported by Office of Naval Research grant #00014-98-1-0597 through the University of North Carolina in Chapel Hill, North Carolina, United States of America. Thus, the United States government has certain rights in the invention.

Technical Field

The present invention relates, in general, to a vapor phase deposition method for the preparation of single walled carbon nanotubes, where the method employs a metal catalyst on a support. More particularly, the present invention relates to an improved method where the support comprises an aerogel, such as an Al_2O_3 aerogel or an Al_2O_3/SiO_2 aerogel, as compared to prior art methods that employed supports that are powders. The improved method results in far higher yields of single walled carbon nanotubes than the prior art methods.

Abbreviations

ASB	aluminum tri-sec-butoxide
AFM	atomic force microscope
$(acac)_2$	bis(acetylacetonato)
cm	centimeter
C	centrigrade
CVD	chemical vapor deposition
EtOH	ethanol
g	gram
kg	kilogram
kV	kilovolt

m	meter
ml	milliliter
MW	molecular weight
MWCNT	multi-walled carbon nanotube
nm	nanometer
psi	pounds per square inch
SEM	scanning electron microscope
SWCNT	single walled carbon nanotube
sccm	standard cubic centimeter per minute
STP	standard temperature and pressue
Tpa	tera pascal
TGA	thermal gravimetric analyzer
TEM	transmission electron microscope

Background of the Invention

Ever since the discovery by lijima in 1991 of the carbon nanotube, it has been one of the most actively studied materials in today's research. See, lijima, Vol. **354**, *Nature*, pp. 56-58 (1991). This active study is not very surprising given the outstanding chemical and physical properties that this material possesses and its potential applications in many different fields.

For example, depending on the number of concentric walls of a graphene sheet and the ways that a graphene sheet is rolled into a cylinder, carbon nanotubes can be conductive behaving like metals or can be semi-conductive. See, Dresselhaus et al., *Science of Fullerenes and Carbon Nanotubes*, Academic Press, San Diego (1996).

Furthermore, experiments have shown that individual carbon nanotubes can behave as quantum wires and can even be made into room temperature transistors. See, Tans et al., Vol. **386**, *Nature*, pp. 474-477 (1997) vis-à-vis quantum wires and Tans et al., Vol. **393**, *Nature*, pp. 49-52 (1998) vis-à-vis transistors.

In addition, carbon nanotubes have been shown to possess superior mechanical properties and chemical stability. Experimental measurements of

carbon nanotubes for Young's moduli by AFM and for thermal vibrations afforded respective values of 1.3 Tpa and 1.8 Tpa, which are higher than the values for any other known material. See, Wong et al. Vol. **277**, *Science*, pp. 1971-1975 (1997) vis-à-vis AFM and Treacy et al., Vol. **381**, *Nature*, pp. 678-680 (1996) vis-à-vis thermal vibrations.

Consequently, the chemical stability, the superior mechanical properties, the ballistic transport property of metallic-like behavior, and the rich variation in electronic properties due to different helicities make carbon nanotubes ideal candidates for high strength composite materials, and for interconnections and functional devices in molecular electronics.

Although carbon nanotube materials possess many unique and technically important properties, lack of a way to produce a sufficient amount of materials has limited not only the study of the fundamental properties but also the development of more practical applications. The discovery of a low cost, high yield method for preparation of SWCNT material will certainly solve one of the biggest problems facing this field in the past and open new opportunities for a wide variety of applications.

Currently, carbon nanotubes are synthesized by three different techniques: (1) arc discharge between two graphite electrodes, (2) CVD through catalytic decomposition of a hydrocarbon or of CO, and (3) laser evaporation of the carbon target. With respect to CVD, see, International Publication No. WO 89/07163 to Synder et al.; U.S. Patent No. 4,663,230 (issued in 1987) to Tennent et al.; M. Terrones et al., *Nature* **388**, 52-55 (1997); Z. F. Ren et al., *Science* **282**, 1105-1107 (1998); J. Kong, A. Cassell, and H. Dai, *Chemical Physics Letters* **292**, 4-6 (1998); J. H. Hafner et al., *Chemical Physics Letters* **296**, 195-202 (1998); E. Flahaut et al., *Chemical Physics Letters* **300**, 236-242 (1999); S. S. Fan et al., *Science* **283**, 512-514 (1999); H. J. Dai et al., *Chemical Physics Letters* **260**, 471-475 (1996); H. M. Cheng et al., *Applied Physics Letters* **72**, 3282-3284 (1998); and A. M. Cassell, J. A. Raymakers, J. Kong, and H. J. Dai, *Journal of Physical Chemistry B* **103**, 6484-6492 (1999).

Both the laser method and the arc method yield high quality SWCNTs. However, both techniques suffer from the problem that it is hard to increase the production volume of the nanotube materials from laboratory scale to industrial scale.

On the other hand, based on published reports, the CVD method appears to represent the best hope for large scale production of nanotube materials. This method has been reported in the 1980's (by Tennent et al. in U.S. Patent No. 4,663,230 (issued in 1987) and by M. S. Dresselhaus, G. Dresselhaus, K. Sugihara, I. L. Spain, and H. A. Goldberg in *Graphite Fibers and Filaments*. M. Cardona, et al., Eds., Springer Series in Materials Science 5 Springer-Verlag, New York (1988) vol. 5) for preparation of various carbon materials such as carbon fibers and multi-walled carbon nanotubes with a yield higher and on a scale larger than those reported for the laser method and the arc method.

More recently in the 1990's, there have been reports of SWCNT preparation by CVD (carbon monoxide or methane) and reports of SWCNT preparation, mixed with a substantial amount of MWCNT preparation, by CVD (benzene or ethylene). With respect to carbon monoxide CVD, see, H. J. Dai et al., *Chemical Physics Letters* **260**, 471-475 (1996) and P. Nikolaev et al., *Chemical Physics Letters* **313**, 91 (1999). With respect to methane CVD, see, A. M. Cassell, J. A. Raymakers, J. Kong, and H. J. Dai, "Large Scale CVD Synthesis of Single-Walled Carbon Nanotubes", *Journal of Physical Chemistry B* **103**, 6484-6492 (1999) and E. Flahaut et al., *Chemical Physics Letters* **300**, 236-242 (1999). With respect to benzene CVD, see, H. M. Cheng et al., *Applied Physics Letters* **72**, 3282-3284 (1998). With respect to ethylyne CVD, see J. H. Hafner et al., *Chemical Physics Letters* **296**, 195-202 (1998). Hence, although the report for ethylene and the report for benzene each mentions SWCNTs, they have the drawback that they are always mixed with a substantial amount of MWCNTs.

Among these reported CVD methods, only the methane CVD method has been reported to produce high purity and high quality SWCNT materials. However, the reported yield of the methane CVD method is low, with the best

results so far giving a total yield of 40% based on the weight of catalyst for a reaction time of 10 to 45 minutes, where the catalyst was supported on Al_2O_3 powder or on Al_2O_3/SiO_2 powder and the catalyst/support had a surface area of about 100 m^2/g. See, Cassell et al., supra.

Thus, a CVD method that affords high quality SWCNTs in a far higher yield (such as at least about 100% for a reaction time of about 30 minutes) would be desirable.

Summary and Objects of the Invention

Accordingly, the present invention provides vapor phase method that employs a metal catalyst supported on an aerogel, for instance on Al_2O_3 aerogel and/or on Al_2O_3/SiO_2 aerogel. The catalyst/support employed in the present invention was made by solvent-gel synthesis with subsequent removal of the liquid solvent by drying selected from the group consisting of supercritical drying, freeze drying and combinations thereof, with supercritical drying being preferred. The inventive method involves vapor phase depositing on the catalyst/support a carbon-containing compound. The compound should have a molecular weight of 28 or less, and if the compound has a higher molecular weight, then the compound should be mixed with H_2. The vapor phase depositing is with sufficient heat for a sufficient time, in order to produce SWCNTs on the aerogel supported catalyst. Then, the SWCNTs may, if desired, be removed from the aerogel supported catalyst. Typically, the SWCNTs are produced in high yield, for instance, about 100% or greater, based on the weight of the catalyst.

Thus, it is an object of the invention that in a preferred embodiment SWCNTs are obtained in high yields heretofore unobtainable. This yield is far higher than that of the prior art CVD method, which resulted, at best, in a yield of about 40% based on the weight of the catalyst.

Hence, it is an advantage that this inventive discovery affords a way to prepare SWCNT materials on a large scale, i.e., industrial scale, with low cost.

Some of the objects and advantages of the invention having been stated, other objects will become evident as the description proceeds, when

taken in connection with the laboratory examples and drawings described below.

Brief Description of the Drawings

Figure 1 is a graph showing typical TGA yield curves for (a) as-prepared and (b) purified SWCNT materials in air, made in accordance with the inventive method.

Figure 2 is a graph showing weight gain versus reaction time at 900°C with a methane flow at 1158 sccm for a SWCNT material prepared by the inventive method.

Figures 3a and 3b are, respectively, photographs taken through a microscope showing (a) a SEM image and (b) a TEM image of a SWCNT sample prepared by the inventive method on an Al_2O_3 aerogel supported Fe/Mo catalyst. The sample was prepared at about 900°C under a CH4 flow. The flow rate was 1158 sccm. The reaction time was 30 minutes.

Detailed Description of the Invention

The present invention provides single walled carbon nanotubes using a novel vapor phase method in which a particular catalyst/support is employed in deposition of a carbon-containing compound. In a preferred embodiment, the present invention provides a dramatic increase in the yield of single walled carbon nanotubes as compared to the prior art method that uses powder for a support.

By the term single walled carbon nanotubes is meant what is conventionally known in the art. Moreover, with the inventive method, it is not intended to exclude that a minor amount, for instance < 1%, of multi-walled carbon nanotubes may be concurrently produced.

A suitable carbon-containing compound may be one that is vapor at STP or may be one that is capable of being converted into vapor at reaction conditions. Preferably, the compound is one that has a molecular weight of 28 or less. Examples are CO, CH_4, and combinations thereof. If the compound has a molecular weight greater than 28, for instance benzene (MW = 78) or

ethylene (MW = 30), then the compound should be admixed with H_2, for instance, 50% by volume H_2.

To effect the preferred embodiment of a high yield of about 100% or more, a sufficient flow rate of the carbon-containing compound should be employed, and may range from about 900 sccm to about 1300 sccm.

A sufficient time may range from about 0.25 hours to about 7 hours. A sufficient temperature may range from about 750°C to about 1000°C. The yield may be about 200%, about 300%, or even higher.

A suitable catalyst is any metal catalyst known in the art for making nanotubes. A preferred metal catalyst may be Fe/Mo, Fe/Pt, and combinations thereof. A suitable support is any aerogel as that term is conventionally adopted in the art to mean a gel with air as dispersing agent prepared by drying. The aerogel support could be a powdered support converted to an aerogel by known methods. As discussed in more detail below, the drying may be supercritical drying or may be freeze drying, but it is not intended to include drying that results in a xerogel. A preferred aerogel support may be Al_2O_3 aerogel support, Al_2O_3/SiO_2 aerogel support, and combinations thereof.

As shown in Figure 1, the yield of the SWCNT material was measured by heating up the prepared SWCNT material under flowing air in a TGA. The total yield of SWCNT material, which yield is shown on the vertical axis as a % weight gain, was calculated by the weight loss between 300°C and 700°C, which temperature is shown on the horizontal axis, where the SWCNT material burned in air, divided by the weight left at 700°C, which was presumably the weight of the catalyst and support materials.

Purification of the material prepared in the inventive method was also studied. Because of the highly amorphous nature of the aerogel support prepared in the inventive method, removing the catalyst and support from the SWCNT material turned out to be quite easy. The support can be removed by stirring in dilute HF acid, refluxing in another dilute acid (such as HNO_3), or refluxing in dilute base, such as NaOH solution. Figure 1 shows the TGA result of the material refluxed in 2.6 M HNO_3 for about 4 hours, followed by filtration.

As shown in Figure 2, for a typical growth time of about 60 minutes at about 900°C, the average yield using this catalyst/support was about 200%. The maximum yield (weight gain) was found to be about 600% for about 6.5 hours of growth. The inventive method showed a yield of significantly better than the values previously reported values by A. M. Cassell, J. A. Raymakers, J. Kong, H. J. Dai, *Journal of Physical Chemistry B* **103**, 6484-6492 (1999) Kong, Cassell, and Dai, *Chemical Physics Letters* **292**, 4-6 (1998).

As shown in Figures 3a and 3b, the quality of the prepared SWCNT was characterized by SEM imaging and TEM imaging.

More particularly, as depicted in Figure 3a, the SEM image of the as-prepared SWCNT material showed a tangled web-like network of very clean fibers. The diameters of the fibers appeared to be in the range of about 10 to about 20 nanometers. It is noted that the SEM image was of as-grown materials; no purification was performed before the imaging.

Furthermore, as depicted in Figure 3b, the TEM image of the SWCNT material showed that the fibers observed in the SEM image were actually bundles of single walled carbon nanotubes. The diameters of the nanotubes measured from the high resolution TEM images were between about 0.9 and about 2.7 nm.

Both the SEM and the TEM images showed the SWCNT materials possessed characteristics similar to those of high quality single walled carbon nanotube materials prepared in the laser method (see, A. Thess et al., *Science* **273**, 483-487 (1996) and T. Guo, P. Nikolaev, A. Thess, D. T. Colbert, and R. E. Smalley, *Chemical Physics Letters* **243**, 49-54 (1995)) and the arc method (see, M. Wang, X.L. Zhao, M. Ohkohchi, and Y. Ando, *Fullerene Science & Technology* **4**, 1027-1039 (1996) and C. Journet et al., *Nature* **388**, 756-758 (1997)).

The fact that the SEM image showed only nanotubes, but no amorphous carbon overcoat, indicated that the catalyst/support surface was substantially fully covered with nanotube materials. However in the TEM image, for samples with a weight gain, i.e., yield, higher than about 300%, an amorphous carbon

overcoat was observed, which probably can be eliminated during production of SWCNTs and/or removed after production of SWCNTs.

Moreover, the inventive method reflects that a drying process of the wet gel, as discussed below in the laboratory examples, is a necessary step in preparing the high performance catalysts on aerogel supports, as employed in the inventive method. The drying may be achieved by supercritical drying, such as by CO_2 supercritical drying, or by ethanol supercritical drying, or alternatively, may be achieved by freeze drying, such as by freeze drying using water, and combinations thereof.

However, it is not intended to include drying that results in a xerogel. Fricke, *Aerogels,* Springer-Verlag, Berlin, Heidelberg, New York, Tokyo (1986) and N. Husing, U. Schubert, *Angew. Chem. Int. Ed.* **37**, 22-45 (1998) discuss that merely evaporating the liquid solvent at ambient conditions (i.e., about STP) would cause the gel to shrink due to the collapse of the porous structures by the strong forces from surface tension at the liquid/gas interfaces within the pores in the gel, and this shrinkage would significantly reduce the total surface area and pore volume of the dried material, which is normally called xerogel.

On the other hand, in the supercritical drying process, which is performed at a temperature well above STP and should also be at a pressure well above STP, the liquid solvent in the wet gel is put into the supercritical state, for instance, under a carbon dioxide blanket. Therefore, there are substantially no liquid/gas interfaces in the pores during drying. The original porous structure in the wet gel is thus substantially maintained in the resultant dried catalysts/aerogels.

Also, as more and more nanotubes were grown on the surface of the aerogel supported catalyst, the diffusion of the carbon-containing compound, i.e., methane or carbon monoxide in the examples below, to the catalyst/support became more difficult. Furthermore, since as noted in the above discussion of Figures 3a and 3b, an amorphous carbon deposition was observed on the nanotubes at a longer growth time, this carbon overcoat probably further reduced the diffusion rate of the carbon-containing compound

to the catalyst/support. This overcoat would explain why the growth rate slowed down versus time as shown in Figure 2.

In summary, discovered was a new method employing a form of catalyst/support that can be used in a vapor phase deposition method to prepare single walled carbon nanotubes, preferably in yields greater than those obtainable by prior art processes. The yield was typically improved by at least a factor of 2.5 and often 5, compared with a similar catalyst supported on Al_2O_3 powder.

Laboratory Examples

Materials.

All materials used in the laboratory examples were research grade materials purchased from different suppliers.

Aluminum tri-sec-butoxide (abbreviated below as ASB), $Fe_2(SO_4)_3.4H_2O$, and bis(actylacetonato)dioxomolybdenum (abbreviated below as $MoO_2(acac)_2$) were purchased from Sigma/Aldrich Chemicals.

Reagent grade nitric acid, ammonium hydroxide, and ethanol were purchased from VWR Scientific Products.

High purity methane, carbon dioxide, and hydrogen were supplied by National Welders Inc.

EXAMPLE I

Catalyst/Support Preparation.

Catalysts/supports were prepared using the solvent-gel technique , as reported in D. J. Suh and J.T. Park, *Chemistry of Materials* **9**, 1903-1905 (1997) followed by supercritical drying. Optionally, some were dried by freeze drying.

In a typical experiment, 23 g of ASB were dissolved in 200 ml of ethanol as the liquid solvent in a round bottom flask under reflux conditions. Then, 0.1 ml of concentrated HNO_3, diluted with 1 ml of water and 50 ml of ethanol, was added into the mixture.

The resultant was refluxed for 2 hours, until a clear solution was formed, followed by adding 1.38 g of $Fe_2(SO_4)_3.4H_2O$ and 0.38 g of $MoO_2(acac)_2$ into the mixture. The amount of Fe and Mo were chosen so that the molar ratio of Mo:Fe:Al = 0.16:1:16. After refluxing for 2 more hours, the mixture was cooled to room temperature and then 5 ml of concentrated NH_4OH, diluted with 5 ml of water, was added into the mixture under vigorous stirring, in order to enhance that the dissolved metal salts would form nm sized hydroxide particles and would attach to the aerogel. Within a few minutes, a gel formed.

The resultant was left to age for about 10 hours before the supercritical drying step was performed under the following conditions.

First, the catalyst/support wet gel was sealed in a high-pressure container, which was then cooled to about 0°C and pressurized to fill the container with liquid CO_2, at about 830 psi (about 59.4 kg/cm^2). A solvent exchange step followed, in order to exchange the ethanol liquid solvent in the gel with liquid CO_2, by flushing the container with liquid CO_2 a few times.

Then, the container was warmed up to between about 50°C and about 200°C, which is above the critical temperature (31°C) of CO_2, and the pressure was kept between about 1500 psi and 2500 psi (between about 106.4 kg/cm^2 and 176.8 kg/cm^2), which is above the critical pressure (1050 psi, 74.8 kg/cm^2) of CO_2. The system was held at these conditions for a short time before the pressure was slowly reduced while the temperature was kept the same.

Finally, the temperature was reduced to room temperature. Then, each catalyst (in metal hydroxide form) on aerogel support was calcined at 500° C for 30 minutes, to effect conversion to the metal oxide form. Then before being used for SWCNT growth, conversion to the metal form was effected by reduction under H_2 for 30 minutes at 900° C. The pressure at that stage was about 830 psi (about 59.4 kg/cm^2). Each catalyst/support prepared this way was a catalyst supported on a highly porous, very fine, free-flowing aerogel with a surface area of from about 500 m^2/g to about 600 m^2/g.

Alternatively, instead of CO_2, some samples were supercritically dried with ethanol or dried with freeze drying as follows.

Ethanol supercritical drying: A 100 ml high pressure and high temperature container was used. At least 35 ml of the wet gel was added in the container. Before heating, N_2 was used to flush the system to drive the air out. Then the whole system was sealed and heating was started. After the temperature reached 260°C, the system was maintained at that temperature for about 30 minutes before the EtOH was released slowly. The releasing process took about 15 minutes. The, the system was cooled down gradually and the aerogel supported catalyst taken out. The yield of nanotubes for this was similar to the one dried with CO_2 .

Freeze drying: The ethanol in the wet gel was replaced by water through solvent exchange. Then, the sample was frozen with liquid nitrogen and put in a freeze dryer (Freezone Plus 6, Labconco, Kansas City, Missouri, United States of America). It took a few days to dry the sample totally and the yield of this was lower than the one dried with CO_2.

SWCNT Growth.

SWCNTs were prepared in a simple vapor phase deposition setup made of a tube furnace and gas flow control units. In a typical growth experiment, about 50 mg of a catalyst/support sample were put into an alumina boat inside a quartz tube. Each sample was individually heated to reaction temperature, under an Ar flow at a flow rate of about 100 sccm, and then, the Ar was switched to H_2 (about 100 sccm flow rate) for 30 minutes, before switching to a methane flow (about 1000 sccm) for 30 minutes. An individual sample was heated for each temperature of about 800°C, about 850°C, about 900°C, and about 950°C.

The reaction was carried out for the desired time before the methane flow was turned off and the Ar flow turned on and the temperature reduced to room temperature. Each resultant was then weighed and characterized.

Characterization.

SWCNT samples were fully characterized using TEM imaging and SEM imaging.

TEM imaging was performed on a Philip CM-12 microscope operating at 100 kV. The samples for TEM imaging were prepared by sonicating about

1 mg of material in 10 ml of methanol for 10 minutes and drying a few drops of the suspension on a holy-carbon grid.

SEM imaging was performed on a Hitachi S-4700 microscope with a beam energy of 4 kV by placing the as-grown materials on conductive carbon tape.

The yield of the SWCNT material with respect to the catalyst was measured on a thermal gravimetric analyzer (model SDT 2960, purchased from TA Instruments) under flowing air with a heating rate of 5°C/minute. The observed yield, measured by TGA, was 100.2% as depicted in Figure 1.

EXAMPLE II

The procedure of Example I was substantially repeated, except this time with a methane flow for about 60 minutes (instead of about 30 minutes) and a temperature of about 900°C (instead of various temperatures of about 800°C about 850°C, about 900°C, and about 950°C) and a flow rate of about 1158 sccm (instead of about 1000 sccm), during SWCNT growth. The yield measured by TGA was about 200%.

EXAMPLE III (COMPARISON)

Also, a catalyst/support made from the same Al_2O_3 wet gel, but dried differently to make xerogel, was compared. The aerogel supported catalyst showed a yield of about 200% of high purity SWCNT under a methane flow at about 900°C for about 60 minutes, as reported by Example I. On the other hand, the xerogel supported catalyst showed a weight gain of <5% under the same conditions.

EXAMPLE IV

The procedure was repeated as per the Fe/Mo catalyst supported on Al_2O_3 aerogel, but this time with the Fe/Mo catalyst instead supported on SiO_2 aerogel prepared by a similar method.

The weight gain of the catalyst on SiO_2 aerogel under the same conditions, about 900°C under a methane flow for about 60 minutes, was almost 10%. Thus, it appears that although a SiO_2 aerogel support works (i.e., about 10%), it is preferred with the inventive method to employ an Al_2O_3 aerogel support or an Al_2O_3/SiO_2 aerogel support to obtain improvements that are far superior (i.e., weight gain of about 100% or greater).

EXAMPLE V

The procedure of Example I was substantially repeated, except this time with CO instead of CH_4. Also, the temperature of the CO flow was about 850°C, with a CO flow rate of about 1200 sccm for about 200 minutes. The result was a yield of about 150%.

EXAMPLE VI

The procedure of Example I was substantially repeated, except this time with Al_2O_3/SiO_2 as the aerogel support, instead of Al_2O_3 as the aerogel support. Substantially the same results were obtained, except there was more amorphous carbon.

EXAMPLE VII (COMPARISON)

It is believed that more amorphous carbon resulted in Example VI since in a comparison, Al_2O_3/SiO_2 aerogel (without any metal catalyst) was tried with methane for 30 minutes at 900°C and this converted the methane to amorphous carbon.

It will be understood that various details of the invention may be changed without departing from the scope of the invention. Furthermore, the foregoing description is for the purpose of illustration only, and not for the purpose of limitation – the invention being defined by the claims.

CLAIMS

What is claimed is:

1. A method of preparing single walled carbon nanotubes comprising depositing a carbon-containing compound under vapor phase conditions onto a supported catalyst comprising a metal catalyst and an aerogel support while heating under reaction conditions of temperature sufficient and time sufficient to form the single walled carbon nanotubes on the aerogel supported catalyst.

2. The method of claim 1, where the carbon-containing compound has a molecular weight of 28 or less.

3. The method of claim 2, where the carbon-containing compound is selected from the group consisting of methane, carbon monoxide, and combinations thereof.

4. The method of claim 1, where the carbon-containing compound has a molecular weight greater than 28 and is mixed with hydrogen.

5. The method of claim 4, where the carbon-containing compound is selected from the group consisting of ethylene, benzene, and combinations thereof.

6. The method of claim 1, where the metal catalyst is selected from the group consisting of Fe/Mo, Fe/Pt, and combinations thereof.

7. The method of claim 1, where the aerogel support is selected from the group consisting of Al_2O_3 aerogel support, Al_2O_3/SiO_2 aerogel support, and combinations thereof.

8. The method of claim 1, where said depositing is with a sufficient flow rate of the carbon-containing compound, with sufficient heat for a sufficient time, to obtain a yield of at least about 100% based on the weight of the catalyst.

9. The method of claim 8, where the sufficient flow rate ranges from about 900 sccm to about 1300 sccm.

10. The method of claim 1, where the aerogel supported catalyst has a surface area of from about 500 m^2/g to about 600 m^2/g.

11. The method of claim 1, where the aerogel supported catalyst has been dried by drying selected from the group consisting of supercritical drying, freeze drying, and combinations thereof.

12. The method of claim 11, where the supercritical drying is selected from the group consisting of CO_2 supercritical drying, ethanol supercritical drying, and combinations thereof.

13. The method of claim 11, where the freeze drying is freeze drying using water.

14. The method of claim 1, where the sufficient heat provides a temperature ranging from about 750°C to about 1000°C .

15. The method of claim 1, where the sufficient time is at least about 0.25 hours.

16. The method of claim 1, where the yield is at least about 100%.

17. The method of claim 1, further including separating the single walled carbon nanotubes from the aerogel supported catalyst.

18. A method of preparing single walled carbon nanotubes comprising depositing a carbon-containing compound under vapor phase conditions onto a supported catalyst comprising a metal catalyst and an aerogel support where the carbon-containing compound is selected from the group consisting of methane, carbon monoxide, and combinations thereof, where the metal catalyst is selected from the group consisting of Fe/Mo, Fe/Pt, and combinations thereof, where the aerogel support is selected from the group consisting of Al_2O_3 aerogel support, Al_2O_3/SiO_2 aerogel support, and combinations thereof, where the aerogel supported catalyst has been dried by drying selected from the group consisting of supercritical drying, freeze drying, and combinations thereof, and where said depositing is with a sufficient flow rate of the carbon-containing compound, with sufficient heat for a sufficient time, to obtain a yield of at least about 100% based on the weight of the catalyst.

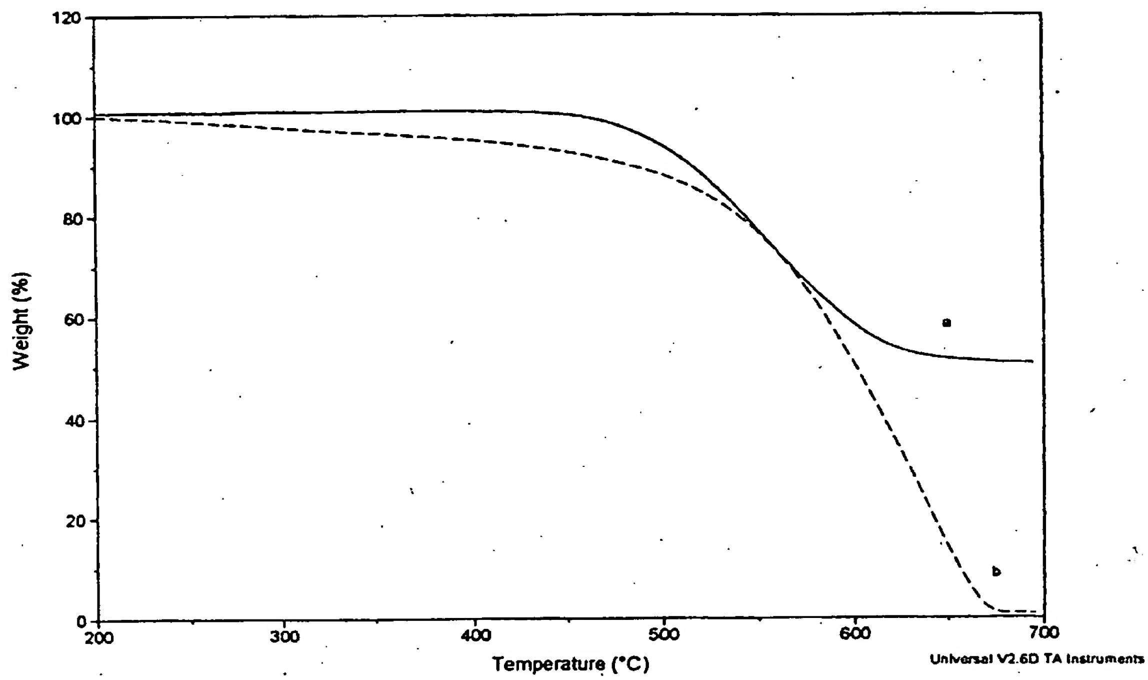


Figure 1. Typical TGA curves for (a) as-prepared and (b) purified SWNT materials in air.

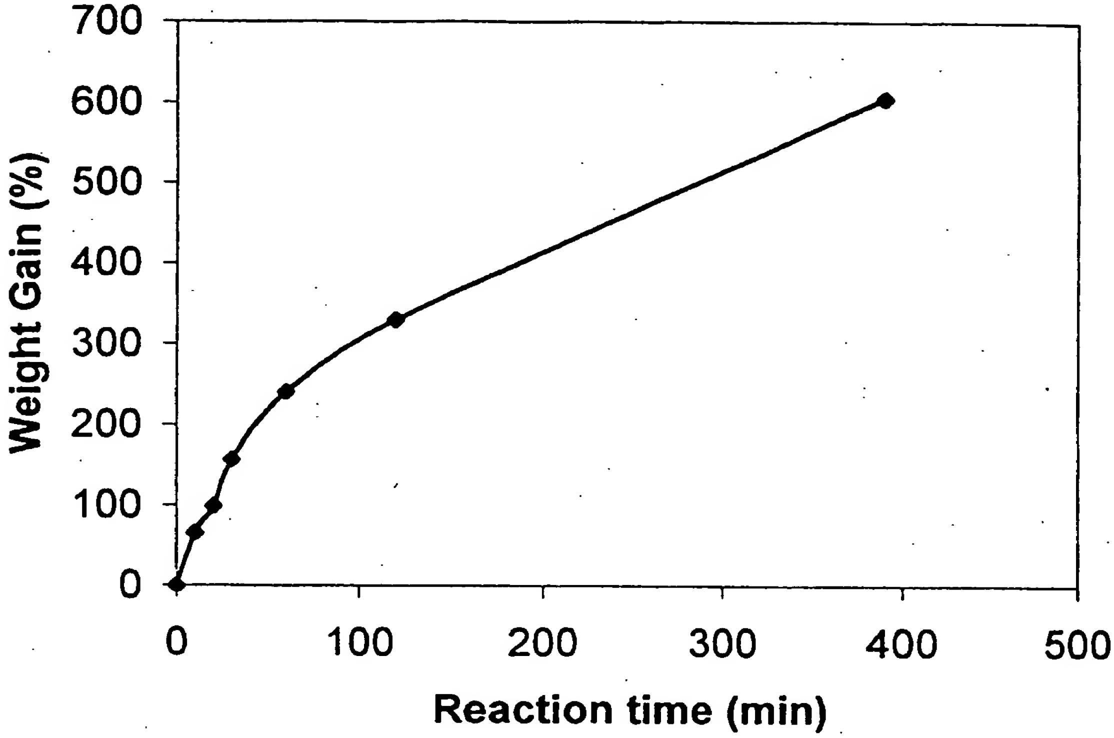
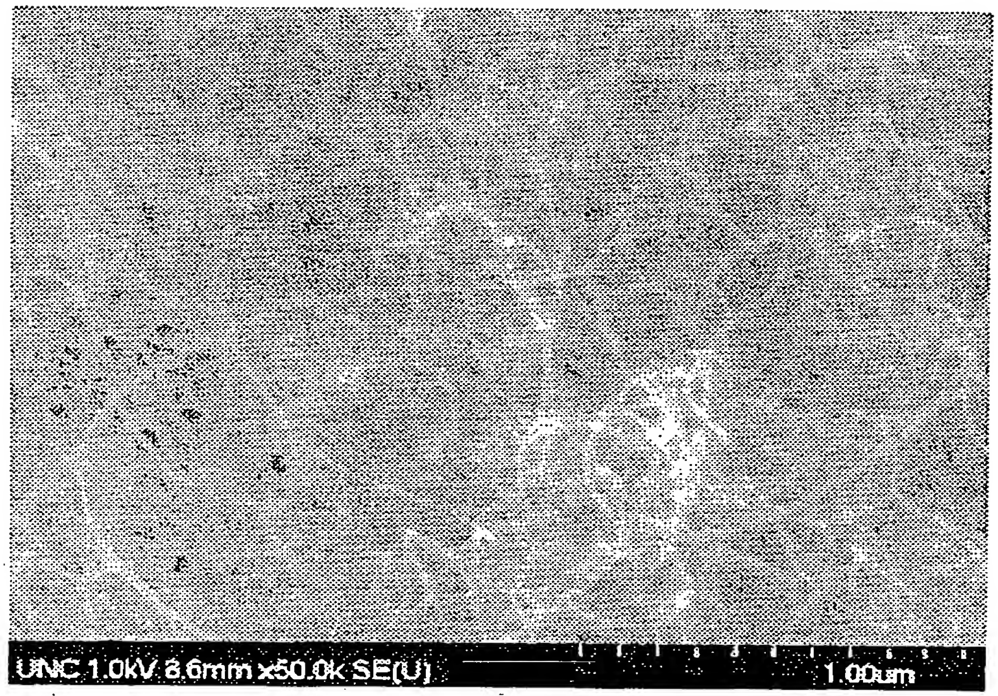


Figure 2: Weight gain vs. reaction time plot at 900°C with methane flow at 1158 sccm.



(a)

FIG. 3(a)

(b)

Fig. 3 (b)

Figure 3. (a) SEM and (b) TEM image of SWNT sample prepared on Al_2O_3 aerogel supported Fe/Mo catalyst. Sample was prepared at 900°C under CH_4 flow. Flow rate is 1158 sccm; Reaction time is 30 min; Yield measured by TGA is 100.2%.